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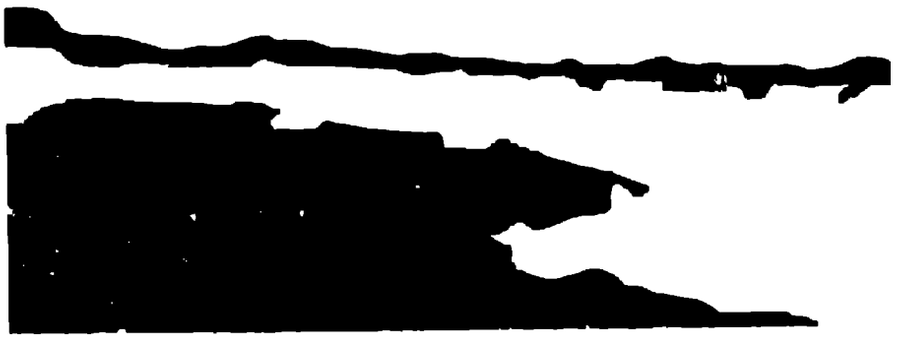

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Magnetic properties of single-crystalline UCu_3Al_2

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Abstract-UCu_3Al_2 crystallizes in an ordered variant of the hexagonal $CaCu_5$ structure. By neutron powder-diffraction, the U atoms were found to occupy the 1a sites, while the 2c sites are occupied by Cu atoms only and a random occupation of the 3g sites by the remaining Cu and Al is found. The magnetic susceptibility, measured on a single crystal grown by the Czochralski tri-arc technique, is found to be maximal within the hexagonal basal plane with a maximum at about 10 K. For fields applied within the basal plane, the magnetization at 4.2 K exhibits a slight S-shape starting slightly below 15 T. No such anomalies are found for fields applied along the c-axis where the magnetic response is found to be much lower. No additional magnetic peaks, which could be related with long-range antiferromagnetic ordering, were detected in the neutron powder-patterns at low temperatures.

I. Introduction

The broad variety of different phenomena found in ternary UTX compounds (T = transition metal, X = element from the p-blocks of the periodic table) have stimulated extensive investigations, mainly on compounds crystallizing in the stoichiometry 1:1:1 [1]. The recent discovery of the heavy-fermion superconductors UNi_2Al_3 [2] and UPd_2Al_3 [3], both crystallizing in a crystal structure related to the hexagonal $CaCu_5$ structure, has focused the attention to compounds forming in this structure or related ones. As to compounds with Cu and Al only the compound $UCu_{3.5}Al_{1.5}$ was reported up to now to crystallize in the $CaCu_5$ structure [4]. Recently, UCu_xAl_{5-x} compounds in a composition range $2.9 < x < 3.5$ were found to synthesize in structures related to the $CaCu_5$ structure [5]. Possible heavy-fermion behaviour in these compounds is indicated by the high C/T-values, which exceed 300 $mJ/molK^2$ at low temperatures [5],[6]. In this paper, we report on the crystal structure and the magnetic properties of single-crystalline UCu_3Al_2.

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II. Sample Preparation and Characterisation

Polycrystalline UCu_3Al_2 was prepared by arc-melting appropriate amounts of the elements with a purity of at least 4N. This ingot was used to prepare a single crystal by means of the Czochralski tri-arc technique. We obtained a bulk rod of approximately 9 g containing single-crystalline parts and parts, in which some twinning within the hexagonal basal plane was found. The single-crystalline parts were extracted from the rod and the quality was checked by X-ray Laue-diffraction. The maximum mass of these crystals was 50 mg. Part of the rod was inspected by microprobe analysis for the composition and possible impurities. An average composition of 16.0%, 51.2% and 32.8% for uranium, copper and aluminium, respectively, was found, which is close to the exact 1:3:2 composition. Furthermore, a small amount (less than 2%) of pure uranium was detected.

In order to resolve the crystal structure, the remaining pieces from the rod were ground and enclosed in a sealed vanadium can under He atmosphere. This can was mounted in the High Intensity Powder Diffractometer (HIPD) at the Manuel Lujan,Jr. Neutron Scattering Center (LANSCE) at Los Alamos. Data were collected at room temperature in six detector banks (+154°, +90° and +40°).

In Fig. 1, the data set of the +154° detector bank is shown. Rietveld refinement of the data of all six detector banks was performed using the Generalized Crystal Structure Analysis System (GSAS) program [7]. It was possible to index the data by the $CaCu_5$ structure (space group: P6/mmm) with random occupation of the 2c and 3g sites by Cu and Al atoms (Fig. 1a). However, the intensities of the

peaks indicated by arrows in Fig. 1 are not fit very well in this structure. The data were found to be represented much better assuming an almost full occupation of the 2c sites by Cu atoms (see table 1), whereas the 3g sites are occupied randomly by the remaining Cu and Al atoms (Fig. 1b). Thus, UCu_3Al_2 has a layered structure of UCu_2 layers separated by $CuAl_2$ layers, a structure also reported for several compounds containing rare earths [8]. The refined structure parameters for the 'ordered' structure at 300 K are listed in table 1. In this structure the reduced χ^2 amounts to 4.7, which is considerably smaller than a value of 14.9 obtained for the disordered structure. At 300 K, the lattice parameters are 514.020(7) and 414.720(6) pm for a and c, respectively.

III. Experimental results and discussion

The magnetization was measured on a single crystal of approximately 40 mg by means of a pendulum magnetometer in fields up to 1.3 T. The magnetic susceptibility is determined as an average of the M/B-values in eight different fields. The temperature dependence of the magnetic susceptibility in the temperature range from 1.8 to 300 K is shown in Fig. 2. A broad maximum at about 10 K is visible for fields applied along the a and b axis (in orthorhombic notation). This maximum is absent for fields applied along the c axis, for which, at higher temperatures, the response is

TABLE 1 Refined structure parameters for UCu_3Al_2

Space group	P6/mmm, Z = 1				
Element	position	x	y	z	fraction
U	1a	0	0	0	1.0000
Cu(1)	2c	1/3	2/3	0	1.0000
Al(1)	2c	1/3	2/3	0	0.0200
Cu(2)	3g	1/2	0	1/2	0.3333
Al(2)	3g	1/2	0	1/2	0.6667
R factors	R_{wp} = 4.99%		R_p = 3.60%		reduced χ^2 = 4.698 %

found to be roughly half of that in basal plane. Above 50 K, all three axes exhibit Curie-Weiss behaviour, which is demonstrated in the $1/\chi$ vs T (inset in Fig. 2). For the a and b axis, similar values for the paramagnetic Curie-temperature θ_p of about -101 K and the effective moment μ_{eff} of about 3.35 μ_B are obtained, whereas those quantities are -314 K and 3.55 μ_B, respectively, for the c axis. The origin of the deviation in the $1/\chi$ vs T curve for the c axis is not yet clear.

On the same crystal, magnetization measurements were performed at 4.2 K in magnetic fields up to 38 T in the Amsterdam High Field Installation. Two kinds of magnetic-field pulses were used: firstly, step-wise pulses indicated by symbols in Fig. 3, in which the magnetic field is kept constant for 80 ms, and secondly, pulses in which the field varies continuously, indicated by the solid line in Fig. 3. Since the data obtained in continuous pulses may contain eddy-current effects, they have been adjusted to the values of the step-wise pulses. The magnetic response of the crystal measured was extremely small, at the border of the sensitivity

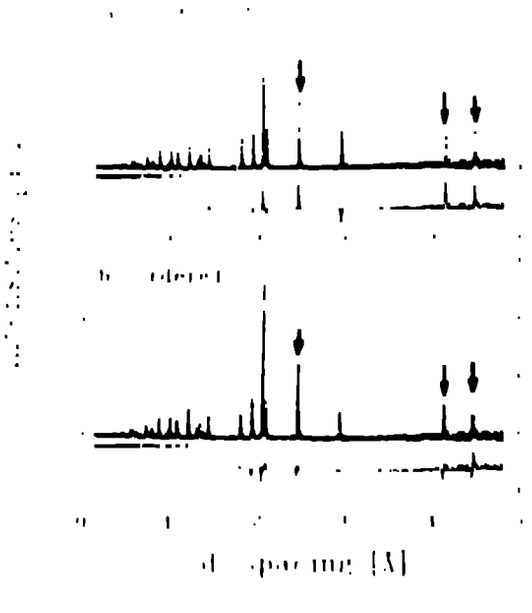

Fig. 1 Portion of the raw data of the 0.15° bank at 100 K. The solid lines represent the calculated intensities for a) random occupation of 2c and 3g sites by the Cu and Al atoms, and b) occupation of the 2c sites by Cu atoms only. The difference of measured and calculated intensities is represented by the solid lines below. Clearly, the peak intensities marked by arrows indicate the latter structure type.

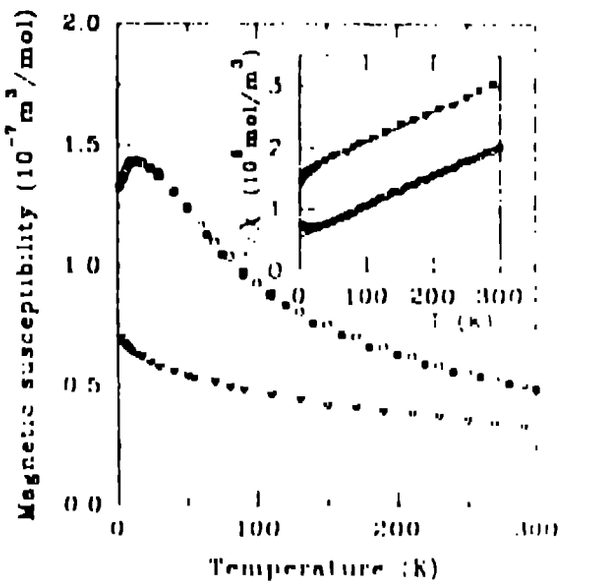

Fig. 2 Temperature dependence of the magnetic susceptibility for magnetic field applied parallel to the a axis (□), b axis (●) and c axis (▽). In the inset, the temperature dependence of the inverse magnetic susceptibility is shown

of the magnetometer. Therefore, the magnetization values obtained in this way were checked in magnetic fields up to 5 T using a SQUID magnetometer. Nevertheless, the absolute values of the magnetization in higher fields may be wrong by about 10%. The overall shape is correct.

The magnetization at 4.2 K, shown in Fig.3, exhibits a slight S-shape starting in fields slightly below 15 T for the *a* and *b* axes yielding values slightly below 1.0 μ_B in 35 T. The continuous pulse reveals a slight tendency toward saturation in the highest fields applied. Although there is some difference in the magnetization values for the *a* and *b* axes, the present data have insufficient precision for a determination of the in-plane anisotropy in UCu_3Al_2. The hard axis in this compound is undoubtedly the *c* axis with an extremely large anisotropy, which is reflected in the much weaker response of about 0.3 μ_B in 35 T. Furthermore, no sign of a S-shape is found for this axis.

In order to determine, whether the maximum in the χ vs T curve is connected with long-range magnetic order, we performed neutron powder-diffraction at 20 K and 4.2 K, which is well above and below the temperature, where the maximum occurs. Therefore, the powder sample, which was used for the crystal-structure determination, was mounted on

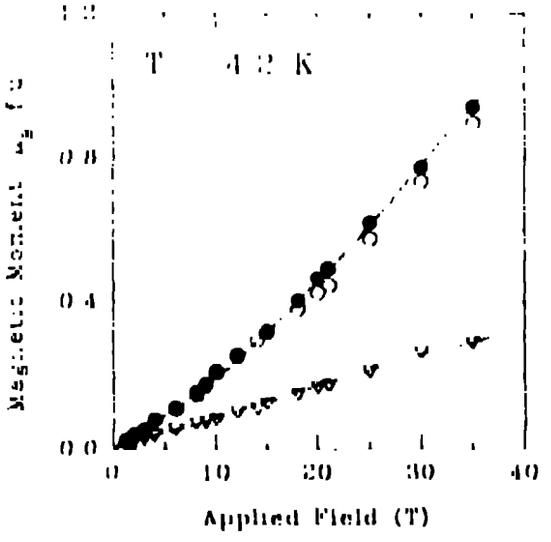

Fig. 3. Field dependence of the magnetization at 4.2 K for *B* parallel to the *a* axis (○), *b* axis (●) and *c* axis (▽). The symbols represent the data obtained in step wise pulses, whereas the solid lines represent the result obtained in field sweeps with continuously changing field.

the BT-4 powder diffractometer at the NIST Research Reactor. A wavelength of 2.35 Å was used and the intensities for scattering angles between 5° and 65° (steps of 0.1°) were detected. Each point was counted for one minute.

No additional peaks, which could be connected with long-range magnetic order, could be resolved in the difference spectrum. Our data indicate that any ordered moment must be less than 0.4 μ_B.

IV. Conclusions

We have reported on a new uranium intermetallic compound UCu_3Al_2, which crystallizes in an ordered variant of the $CaCu_5$ structure. Magnetic measurements reveal strong magnetocrystalline anisotropy with the *c* axis as the hard axis. The response in the basal plane shows anomalies in the field and temperature dependence of the magnetic susceptibility. Although the present neutron-diffraction experiments do not reveal any additional peaks at low temperatures, the ground state is probably antiferromagnetic with rather small moments involved in the ordering. This assumption is supported by the observation, that the specific-heat maximum at 8 K is hardly affected by application of a magnetic field of 5 T [6]. Further support for this assumption is given by compounds with Ga, which crystallize in the same structure, and in which antiferromagnetic long-range order is reported [9].

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